VALIDATION OF METHODS TO MEASURE MASS FLUX OF A GROUNDWATER CONTAMINANT

THESIS

Hyouk Yoon, Captain, ROKA

AFIT/GES/ENV/06M-08

DEPARTMENT OF THE AIR FORCE
AIR UNIVERSITY

AIR FORCE INSTITUTE OF TECHNOLOGY

Wright-Patterson Air Force Base, Ohio

AFIT/GES/ENV/06M-08

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Hyouk Yoon, BS

Captain, Republic of Korea Army

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Hyouk Yoon, BS

Captain, Republic of Korea Army

Approved:

/ SIGNED /	4 Mar 06
Dr. Mark N. Goltz (Chairman)	date
/ SIGNED /	4 Mar 06
Dr. Carl G. Enfield	date
/ SIGNED /	4 Mar 06
Dr. Junqi Huang	date

AFIT/GES/ENV/06M-08

Abstract

Recently, a number of methods have been developed and subsequently applied to measure contaminant mass flux in groundwater in the field. However, none of these methods has been validated by comparing measured and known fluxes at larger than the laboratory-scale.

Recently, a couple of innovative flux measurement methods, the Tandem Recirculating Well (TRW) and Integral Pumping Test (IPT) methods, have been proposed. The TRW method can measure mass flux integrated over a large subsurface volume without extracting water. The IPT method is a simple and easily applicable method of obtaining volume-integrated flux measurements. In the current study, flux measurements obtained using these two methods are compared with known mass fluxes in a meso-scale three-dimensional artificial aquifer.

The TRW method is applied using two different techniques. One technique is simple and inexpensive, only requiring measurement of heads, while the second technique requires conducting a tracer test. The IPT method requires use of one or more pumping and observation wells in various configurations.

The results of the experiments in the artificial aquifer show that the most expensive technique, the TRW method using tracers, provides the most accurate results (within 15%). The TRW method that relies on head measurements appears not to be a viable flux measurement technique because of the large errors that were observed when applying the technique. The IPT method, although not as accurate as the TRW method

using the tracer technique, does produce relatively accurate results (within 60%). IPT method inaccuracies may be due to the fact that the method assumptions (infinite homogeneous confined aquifer at equilibrium) were not well-approximated in the artificial aquifer. While measured fluxes consistently underestimated the actual flux by at least 36% and as much as 60%, it appears that errors may be reduced when one accounts for potential violations of method assumptions.

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Hyouk Yoon

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VALIDATION OF METHODS TO MEASURE MASS FLUX OF A GROUNDWATER CONTAMINANT

I. Introduction

1.1 Motivation

Groundwater is a critical resource, and groundwater contamination by industrial and agricultural chemicals is an important problem throughout the world. To deal with this problem, many countries are making efforts to clean up the groundwater in their regions and a number of technologies and approaches have been developed and used for remediation of groundwater contamination. Due to time and budget constraints, it is important that the contaminated sites that pose the greatest risk to human health and the environment be cleaned up first. In addition, the most efficient technologies should be employed to clean up contaminated sites. In the past, contaminant concentration has been the key parameter used to help decision makers quantify the risk posed by a contaminated site or the efficiency of a remediation technology. However, in recent years, a number of investigators have proposed using contaminant mass flux rather than concentration as a measure to support remediation decision-making (Einarson and Mackay, 2001; Soga *et al*., 2004; U.S. EPA, 2005).

Mass flux is defined as the mass of contaminant crossing a unit cross sectional area of aquifer per unit time. Quantifying mass flux allows us to: 1) prioritize contaminated groundwater sites for remediation, 2) evaluate the effectiveness of source

removal technologies or natural attenuation, and 3) define a source term for groundwater contaminant transport modeling. The ability to measure the mass flux of contaminant in the subsurface is crucial to our effort to manage groundwater contamination (Einarson and Mackay, 2001).

Over the past several years, researchers have been developing methods to measure contaminant mass flux in groundwater (Bockelmann *et al.*, 2003). These methods include the conventional approach of taking concentration measurements at many points using multilevel sampling wells to estimate flux. Innovative methods include: 1) the integral groundwater investigation method (IGIM) which uses a pump test to measure contaminant flux (Bockelmann *et al*., 2003) and 2) the 'flux meter' method that quantifies flux by using a sorbing permeable media placed in a monitoring well to intercept contaminated groundwater and release resident tracers (Hatfield *et al*., 2004). These methods, however, may be expensive, either because of the need to install numerous monitoring wells (e.g. the multilevel sampling approach and the flux meter technique) or the requirement to extract and manage large volumes of contaminated water (*e.g*. the IGIM).

Kim (2005) recently reviewed mass flux measurement methods and found that an innovative method, known as the tandem recirculating well (TRW) method, which makes use of two re-circulating wells that do not extract groundwater from the subsurface, had potential to accurately measure flux while avoiding the disadvantages of other methods currently in use or under development. The key limitation of the TRW method is that except for the initial study reported by Kim (2005), it is untested. Kim's study validated the TRW method in an artificial aquifer, where a known flux was measured. Two

measurement techniques were used; the multi-dipole technique, which relies on the measurement of drawdown and mounding at each TRW, and the tracer test technique, where a tracer is injected into each TRW to quantify the interflow of water between the two re-circulating wells (Kim, 2005). Kim's studies showed that due to the difficulty measuring the relatively small magnitudes of drawdown and mounding induced by the re-circulating wells in the artificial aquifer, the multi-dipole technique was extremely inaccurate. However, the tracer test technique resulted in relatively accurate flux measurements while avoiding the disadvantages of other flux measurement methods currently in use. Kim's studies were limited to two experiments in the artificial aquifer. The flow rates in the wells and through the artificial aquifer were limited and did not vary significantly in the two experiments. Based on the potential demonstrated in these initial studies, further investigation is clearly warranted.

Another new flux measurement method was recently suggested by Brooks (2005). The new method is a simplification of the IGIM that has been tested at a number of sites in Europe (INCORE, 2003). The new method makes use of integral pump test (IPT) data to directly estimate groundwater Darcy velocity without measuring hydraulic conductivity. Knowing concentration and Darcy velocity, flux can be determined. The method works by measuring the head difference between piezometers and pumping wells as a function of flow in the pumping wells. While this new method has been applied a few times in the field, no study under controlled conditions has been conducted to quantify its accuracy.

1.2 Research Objectives

The ultimate goal of this study is to provide remedial project managers and regulators with an accurate and credible flux measurement tool that they can use as a basis for decision making. The objectives of this particular thesis research are to validate the TRW and IPT methods under various conditions and investigate improvements to the methods that may increase their accuracy. To support the objective, we will attempt to find an answer to the following research questions: how is the accuracy of flux measurement by the TRW method, using either the multi-dipole or tracer technique, affected by: a) the number of tracers, b) flow rate in the TRWs? Similarly, we will attempt to determine how: a) number, b) orientation of pumping and monitoring wells, affect the accuracy of the IPT method. We hypothesize that the operating conditions of the TRW and IPT methods can be optimized to increase the accuracy of the flux measurements. For example, we would imagine that larger flow rates in the TRWs with respect to groundwater flow will improve the accuracy of the multi-dipole approach.

1.3 Research Approach

(1) Conduct a literature review of TRW and IPT methods.

(2) Validate the TRW and IPT flux measurement methods using data obtained from meso-scale artificial aquifer experiments, where actual contaminant flux is known

- using different chemicals (e.g. nitrate, bromide) as tracers for TRWs
- changing the TRW pumping rates and the water flow rate through the aquifer
- using different numbers of pumping wells for the IPT method
- varying the locations and orientations of the IPT pumping wells and monitoring

wells with respect to the regional flow direction

(3) Based on the results of the above experiments, compare the accuracy of the measurement techniques under different conditions.

1.4 Study limitations

- Validation of the TRW and IPT method using an artificial aquifer is limited due to the fact that the aquifer does not truly represent conditions that will be encountered in the field. The artificial aquifer is homogeneous, well-controlled (*e.g.* groundwater gradient is held constant in space and time), and on a relatively small scale in comparison to a natural system.

- Variation of the pumping rates in the TRWs is limited due to equipment limitations in the artificial aquifer.

II. Literature review

2.1 Introduction

In this chapter, we review the literature regarding TRW and IPT methods. After looking at why flux is an important parameter to measure, we investigate in some detail the particulars of the TRW and IPT flux measurement methods, which are the focus of this study.

2.2 Background

The goal of groundwater remediation is to reduce the risk posed to human and environmental receptors by contaminants in the subsurface. Thus, when cleaning up a source of groundwater contamination or evaluating the movement of contaminants in a groundwater plume, our focus should not be on the contaminant concentration; it should be on the rate with which contaminant mass is transported toward receptors (i.e. the contaminant mass flux). Einarson and Mackay (2001) showed how the risk, which is a function of the contaminant concentration at a receptor, is related to the flux of contaminant. Considering the example of a contaminant plume being captured by a water supply well (Figure 1), Einarson and Mackay (2001) showed that the contaminant concentration (C_{sw}) in a downgradient water supply well, pumping at rate Q_{sw} can be calculated as:

$$C_{sw} = M_f \times A / Q_{sw} \qquad (1)$$

where M_f is the contaminant mass flux[$ML^{-2}T$] emanating from a contaminant source whose plume is captured by the supply well and A[L^2] is area of the capture zone

orthogonal to the groundwater flow direction that is captured by well.

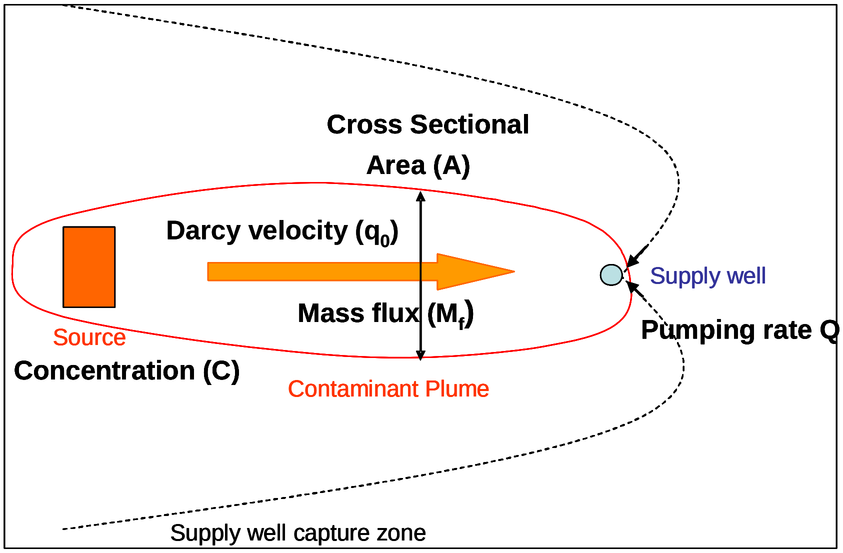

Figure 1. Plan view of a contaminated site (Einarson and Mackay, 2001)

M_f can be obtained from equation (2):

$$M_f = q \times C \qquad (2)$$

where q is the Darcy velocity of the groundwater [L/T] and C [ML^{-3}] is the contaminant concentration emanting from the source area. As shown in equation (2), contaminant concentration, C, is only one component of mass flux. Even though the concentration of contaminant leaving the source area is high, if the Darcy velocity is small, the impact of the source on the downgradient water supply well will be small. As described above, it is contaminant mass flux, rather than contaminant concentration, that is key in determining the risk posed by a contaminant source and plume. To measure the contaminant mass flux, the following methods are in use or being developed: (1) the transect method (Borden *et al.*, 1997), (2) the passive flux meter (PFM) method (Hatfield

et al., 2004), (3) the integral groundwater investigation method(IGIM) (Bockelmann *et al*., 2003), (4) the integral pump test (IPT) method, which is a modified version of the IGIM, proposed by Brooks (2005), and (5) the tandem recirculating well (TRW) method (Kim, 2005; Huang *et al*., 2005).

2.3 Tandem recirculating well (TRW) method

TRWs consist of two pumping wells, with each well having an extraction and injection screen. One well operates in an upflow mode, the other in a downflow mode, so that water recirculates between the two wells without being brought to the surface (see Figure 2).

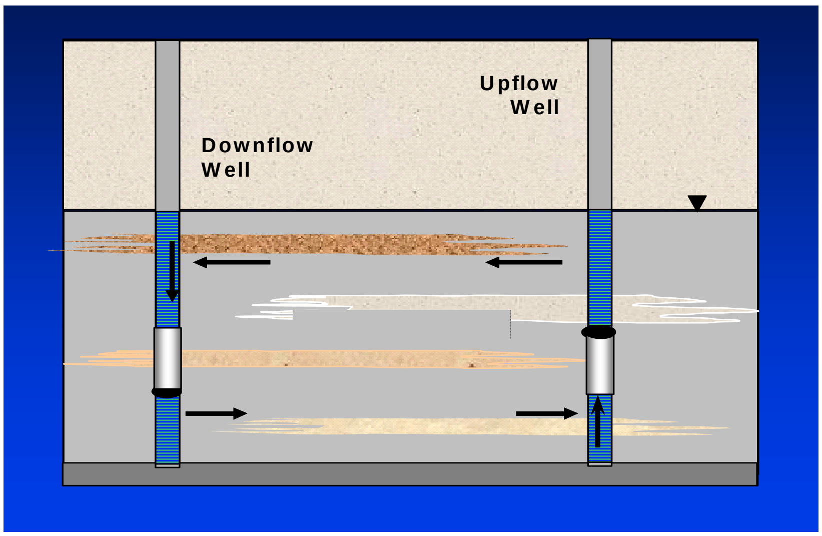

Figure 2. Tandem Recirculating Wells (TRWs)

While TRWs have been applied in the field for contaminant plume cleanup (McCarty *et al*., 1998), and TRW flow models are available (Gandhi *et al.*, 2002), TRWs have not been used in the past for flux measurement. Kim (2005) and Huang et al. (2005) proposed an innovative approach to measure flux by operating TRWs. Since

contaminant mass flux can be calculated as the product of the groundwater Darcy velocity (q_0) and contaminant concentration (C), and, by Darcy's Law, Darcy velocity is the product of hydraulic gradient (i) and hydraulic conductivity (K), the following equation can be used to calculate contaminant mass flux (M_f):

$$M_f = K \times i \times C \tag{3}$$

The TRW method involves individually measuring K, i, and C in order to determine contaminant mass flux. Hydraulic gradient may be determined by measuring the piezometric surface at the two TRWs, with the pumps turned off, and a third piezometer. Volume-averaged contaminant concentration in the TRWs can be measured by sampling the contaminated water as it flows through the wells. To measure hydraulic conductivity, two techniques, both of which were tested by Kim (2005), were proposed. These two techniques, the multi-dipole technique and the tracer technique, are described in detail below.

2.3.1 Multi-dipole technique to measure hydraulic conductivity

The multi-dipole technique is based on the dipole flow test method to measure hydraulic conductivity developed by Kabala (1993). The dipole flow test involves use of a dual-screen well, with a packer separating the screens. The well is pumped at a constant rate, with water flowing in a downward direction; that is, water is extracted from the aquifer into the well through the upper screen and injected into the aquifer through the lower screen. From transient or steady-state drawdown measurements at the upper screen, estimates can be obtained for the value of vertical and horizontal hydraulic conductivity (Kabala, 1993).

Goltz *et al*. (2006) presented an analytical equation to calculate drawdown resulting from operation of a TRW system operating in a horizontally infinite aquifer. Using this analytical formula, if the parameters describing the system are known (well pumping rates, hydraulic gradient, the radius and coordinates of the pumping wells, vertical coordinates of the top and bottom screens, and the thickness of the aquifer) the drawdown and mounding of the wells can be measured to allow calculation of hydraulic conductivities using inverse methods.

By operating the TRWs at a series of different flow rates, the drawdown at the downflow well and the mounding at the upflow well can be measured at each flow rate. Then the analytical formula can be applied to obtain the "best" value of hydraulic conductivity that maximizes the objective function:

$$F_{obj} = \frac{1}{1 + \sum_{i=1}^{N} (H_{meas}^{i} - H_{calc}^{i})^2} \tag{4}$$

where H_{meas}^{i} and H_{calc}^{i} indicate the measured and calculated hydraulic heads at the i^{th} flow rate, respectively, and N is the total number of head measurements. The method can be applied assuming isotropic (that is, horizontal and vertical hydraulic conductivities are the same) or anisotropic conductivities. A genetic algorithm (Carroll, 1996) may be used to determine the best value of hydraulic conductivity that maximizes the objective function (see Figure 3). In this algorithm, each individual value is improved genetically over generations.

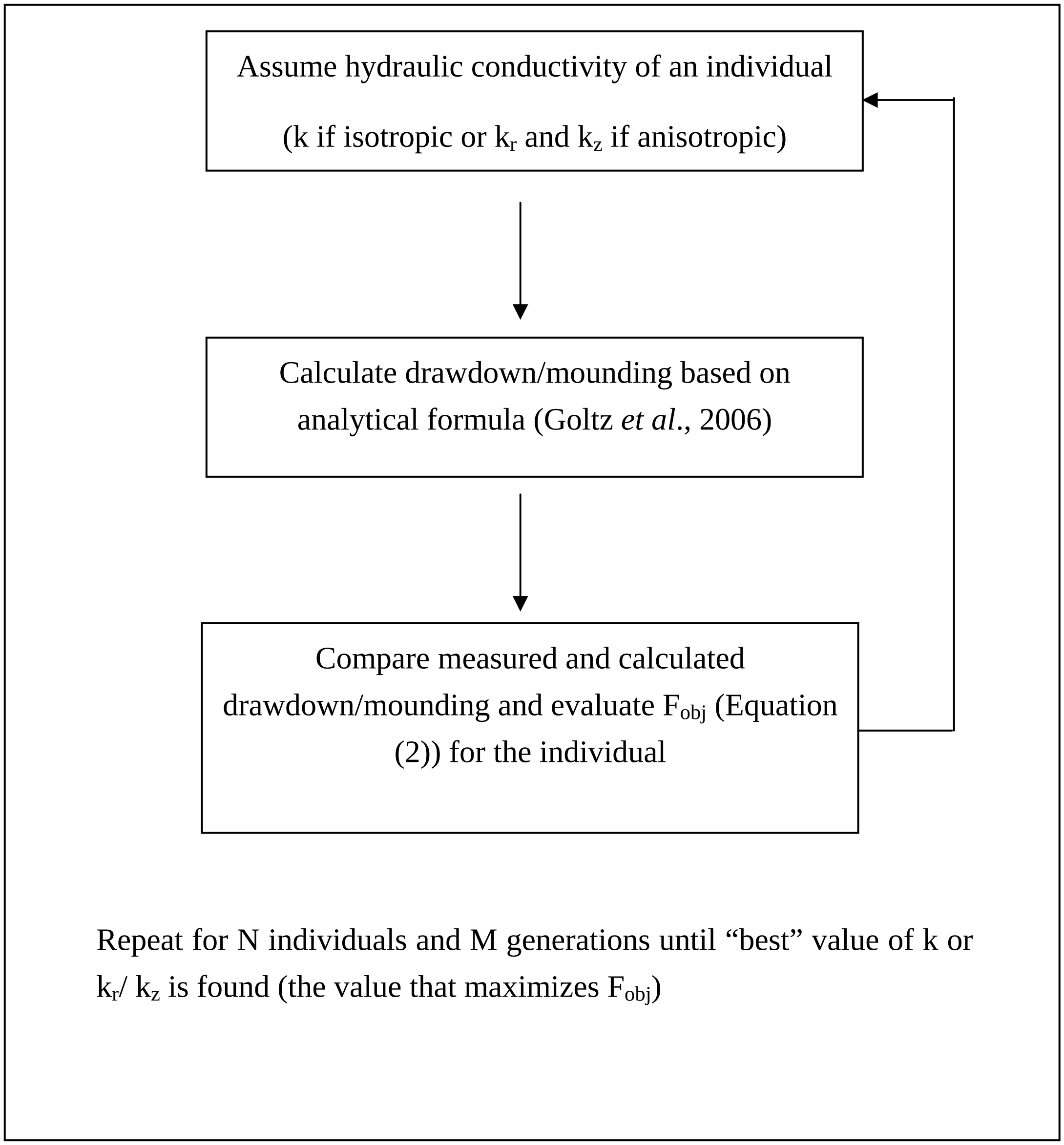

Figure 3. Genetic algorithm procedure

2.3.2 Tracer test technique to measure hydraulic conductivity

The tracer test technique involves operating the TRWs and using tracers to measure hydraulic conductivity.

In Figure 4, I_{ij} represents the fraction of flow entering injection well screen *i* that originated at extraction well screen *j*. As shown in figure 4, tracers can be injected at

the two injection well screens of the two TRWs. If we assume the flow field and the tracer concentrations at the four well screens are at steady-state, the four unknown fractional flows can be obtained using the following four mass balance equations:

$$\begin{aligned} A_1 I_{12} + A_4 I_{42} &= A_2 \\ B_1 I_{12} + B_4 I_{42} &= B_2 \\ A_1 I_{13} + A_4 I_{43} &= A_3 \\ B_1 I_{13} + B_4 I_{43} &= B_3 \end{aligned} \qquad (5)$$

where A_i and B_i are the concentrations of tracers A and B measured at screen i (Huang *et al.*, 2005). As shown by equation (5), these steady-state tracer concentrations are key to determining the four fractional flows accurately. It is potentially difficult to accurately measure the steady-state values of the concentration because there may be concentration fluctuations over time along with random measurement errors. Fortunately, Kim (2005) found that the values of fractional flow obtained from equation (5) were relatively insensitive to the method used in averaging the concentration measurements obtained at the TRWs.

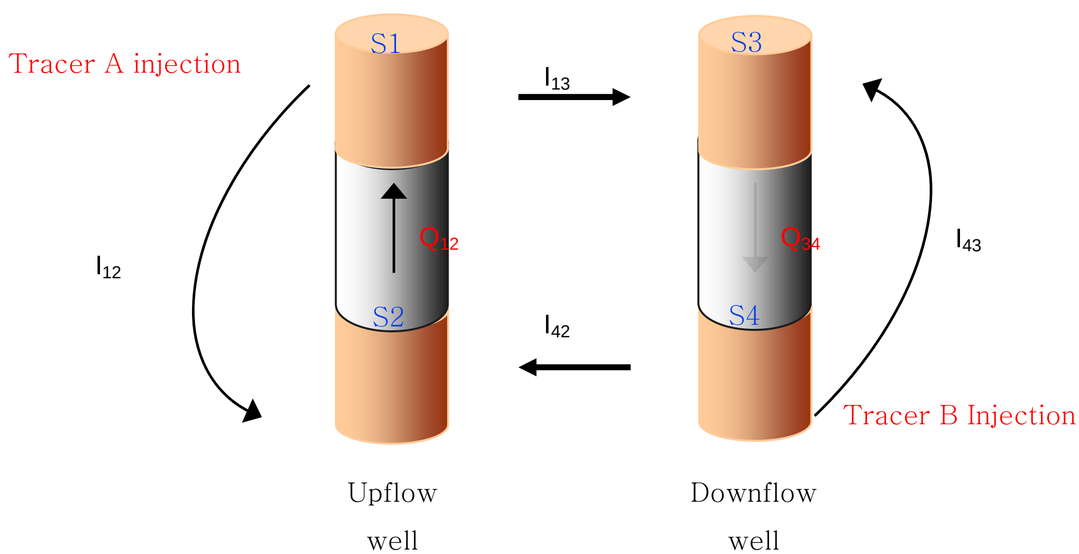

Figure 4. TRW fractional flows and tracer injection screens (Goltz et al., 2004)

With the estimates of the four fractional flows based on the tracer test, inverse numerical modeling can be used to obtain the hydraulic conductivity as follows. Assuming a value of hydraulic conductivity, the three dimensional numerical flow model MODFLOW (Harbaugh and McDonald, 1996) can be used to simulate interflows between the four TRW well screens. The optimal hydraulic conductivity should maximize the following objective function:

$$F_{obj} = \frac{1}{1 + \sum_{i=1}^{N_{inj}} \sum_{j=1}^{N_{ext}} (I_{ij}^{meas} - I_{ij}^{calc})^2} \qquad (6)$$

where I_{ij}^{meas} and I_{ij}^{calc} are the measured and calculated fractional flows, respectively, and N_{inj} and N_{ext} are the number of injection and extraction well screens, respectively.

The method can be applied assuming isotropic (that is, horizontal and vertical hydraulic conductivities are the same) or anisotropic conductivities. A genetic algorithm (Carroll, 1996) may be used to determine the best value of hydraulic conductivity that maximizes the objective function.

2.4 Integral pump test (IPT) method

Brooks (2005) recently suggested the IPT method as a method that can be used to obtain an estimate of contaminant mass flux averaged over a large subsurface volume. The method avoids the data analysis complexities of the IGIM, which requires multiple concentration measurements over time, and unlike the IGIM and TRW techniques, it does not require separate measurements of hydraulic conductivity and hydraulic gradient.

Assuming a homogeneous, isotropic, confined aquifer with uniform thickness under steady-state and uniform flow conditions, the complex potential at some point (x,y)

($W(x,y)$) can be expressed by equation (5) (Javandel, *et al.*, 1984; Christ, J. A. 1997);

$$W(x,y) = \phi(x,y) + i\,\psi(x,y) \qquad (7)$$

Where $\phi\,(x,y)$ is the real velocity potential and $\psi\,(x,y)$ is the imaginary stream function at location (x,y).

The velocity potential (ϕ) at (x, y) is calculated by superposing the potentials due to the uniform regional flow and pumping well sinks (Javandel, *et al.*, 1984):

$$\phi(x,y) = -q_0(x\cos\alpha + y\sin\alpha) + \frac{1}{4\pi B}\sum_{i=1}^{N} Q_i \ln\left[(x-x_i)^2 + (y-y_i)^2\right] + C_1 \qquad (8)$$

where

q_0 = Darcy velocity of uniform regional flow [LT^{-1}]

α = angle between the direction of regional flow and the positive x-axis [-]

B = Aquifer thickness [L]

Q_i = Pumping rate of i^{th} well [L^3T^{-1}]

x_i, y_i = x, y coordinates of i^{th} pumping well, respectively [L]

N = Total number of pumping wells

C_1 = Constant

The hydraulic head (h) is related to the velocity potential by equation (9) (Javandel, *et al.*, 1984):

$$\phi = Kh + C_2 \qquad (9)$$

where K is the hydraulic conductivity and C_2 is a constant.

Combining equations (8) and (9), we obtain:

$$h(x,y) = \frac{-q_0(x\cos\alpha + y\sin\alpha)}{K} + \frac{1}{4\pi BK}\sum_{i=1}^{N} Q_i \ln\left[(x-x_i)^2 + (y-y_i)^2\right] + C \qquad (10)$$

or

$$h(x,y) = -\frac{q_0 B}{T}(x\cos\alpha + y\sin\alpha) + \frac{1}{4\pi T}\sum_{i=1}^{N} Q_i \ln\left[(x-x_i)^2 + (y-y_i)^2\right] + C \qquad (11)$$

where C is a constant and T is the transmissivity $[L^2T^{-1}]$ ($T = KB$).

If we have N pumping wells aligned along the y-axis perpendicular to the regional groundwater flow direction (which is defined as the positive x-direction), and a single observation well on the x-axis at a distance x_{obs} downgradient of the pumping wells (see Figure 5), we can use equations (12) and (13) to calculate the heads at the pumping well located at the origin and the observation well, respectively.

$$h(x_w, 0) = -\frac{q_0 B}{T} x_w + \frac{1}{4\pi T}\sum_{i=1}^{N} Q_i \ln d_{w[i]}{}^2 + C \qquad (12)$$

$$h(x_{obs}, 0) = -\frac{q_0 B}{T} x_{obs} + \frac{1}{4\pi T}\sum_{i=1}^{N} Q_i \ln d_{obs[i]}{}^2 + C \qquad (13)$$

where

x_w = radius of pumping well at the origin [L]

x_{obs} = distance to the observation well along x-axis [L]

$d_{w[i]}$ = distance from the i^{th} pumping well to the pumping well at the origin [L]

$d_{obs[i]}$ = distance from the i^{th} pumping well to the observation well

Subtracting equation (12) from (13):

$$\Delta h = -\frac{q_0 B}{T}\Delta x + \frac{1}{4\pi T}\sum_{i=1}^{N} Q_i \ln \frac{d_{obs[i]}{}^2}{d_{w[i]}{}^2} \qquad (14)$$

where Δx is $x_{obs} - x_w$ and Δh is the difference in heads measured at the pumping well at the origin and the observation well.

We see that when Δh=0, q_0 can be obtained by equation (15):

$$q_0 = \frac{1}{4\pi B\Delta x}\sum_{i=1}^{N} Q_i \ln \frac{d_{obs[i]}^{\ 2}}{d_{w[i]}^{\ 2}} \qquad (15)$$

We also see from equation (14) that if we plot field measurements of Δh as a function of $\sum_{i=1}^{N} Q_i \ln \frac{d_{obs[i]}^{\ 2}}{d_{w[i]}^{\ 2}}$ we should obtain a straight line with slope $1/(4\pi T)$ and intercept $-q_0 B\Delta x/T$. Knowing the Darcy velocity (q_0), contaminant mass flux can be calculated as the product of q_0 and the contaminant concentration measured in the pumping wells.

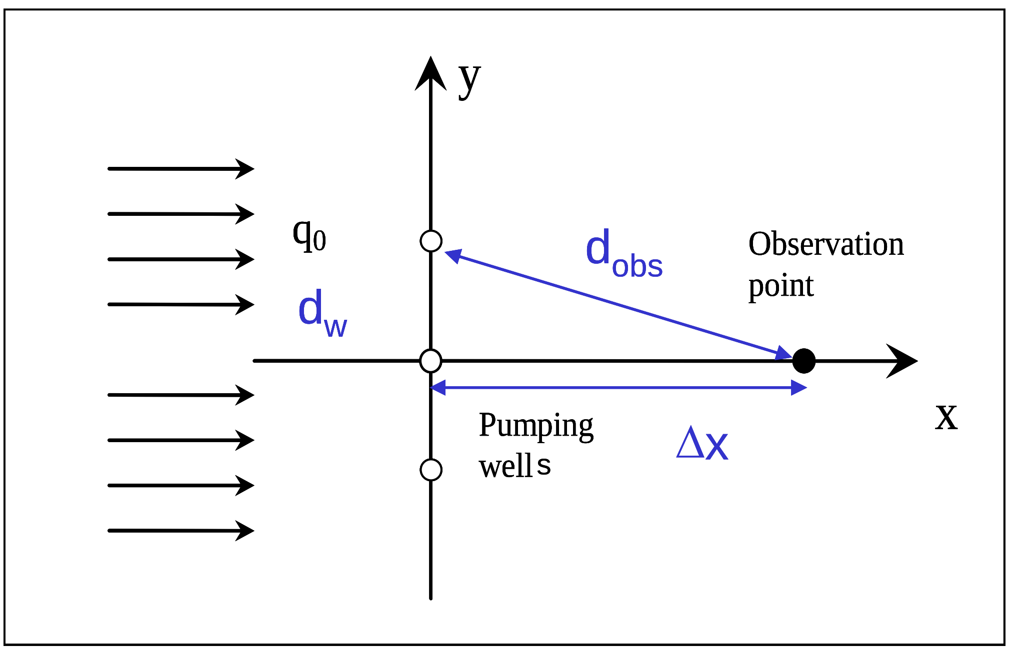

Figure 5. Example of IPT approach with 3 pumping wells and one observation well

III. Methodology

3.1 Introduction

This chapter describes the detailed procedure for measuring mass flux using the TRW and IPT methods. In section 3.2, the artificial aquifer which is used for the flux measurement experiments is described. In section 3.3, experimental conditions and details on the two techniques used in the TRW method, the multi-dipole technique and the tracer test technique, **a**re explained. In section 3.4, experimental conditions and details on the operation of the IPT method are described.

3.2 Artificial aquifer

Evaluation of the TRW and IPT methods was conducted in a meso-scale three dimensional, confined artificial aquifer in Canterbury, New Zealand (Bright *et al*.,2002) (figure 6).

The inner dimensions of the relatively homogeneous sand aquifer are 9.5 *m* long, by 4.7 *m* wide, by 2.6 *m* deep. The aquifer is filled with coarse sand that was dry sieved to fall within the size range 0.6 to 1.2 *mm* in diameter. Constant-head tanks at the aquifer's upstream and downstream ends are used to control the hydraulic gradient. The bottom and sides of the aquifer are no-flow boundaries lined with impermeable butyl rubber.

As shown in Figure 6, there are 45 wells installed on a l *m* by 1 *m* grid, with 9 columns and 5 rows. Each well is a *2.5 cm* diameter tube extending to the bottom of the aquifer. Most of the wells have four sampling ports at depths of 0.4 *m*, 1.0 *m*, 1.6 *m*, and

2.2 *m* below the top of the aquifer, with two wells having seven sampling points. Each sampling port consists of a 7.5 cm long section of well screen with a Teflon sample tube extending from the sampling depth to an automatic sample collector (Bright *et al*., 2002; Kim, 2005). In the TRW and IPT method evaluations, flux of chloride, which is naturally present in the water, was measured.

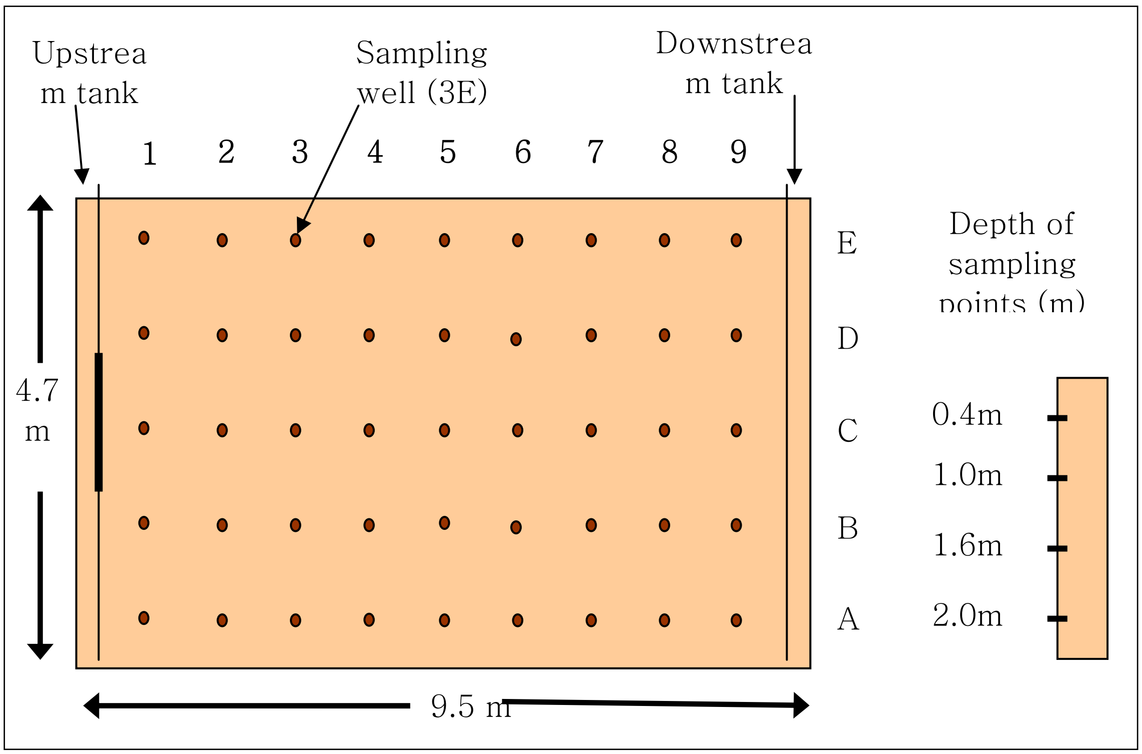

Figure 6. Plan and vertical views of sampling wells in the artificial aquifer (Bright et al., 2002)

3.3 TRW experiment

3.3.1 TRW installation and operation

A TRW well pair was installed in the artificial aquifer at locations 7B and 7D (Figure 7, the upflow TRW at 7D and the downflow at 7B). Water containing chloride

as a model contaminant was continuously input at the upstream tank. The concentration of chloride was measured at the two TRWs and found to average 10.48 mg/L. The water levels were measured at two piezometers, upgradient and downgradient, which were separated by 9.099m to calculate the hydraulic gradient.

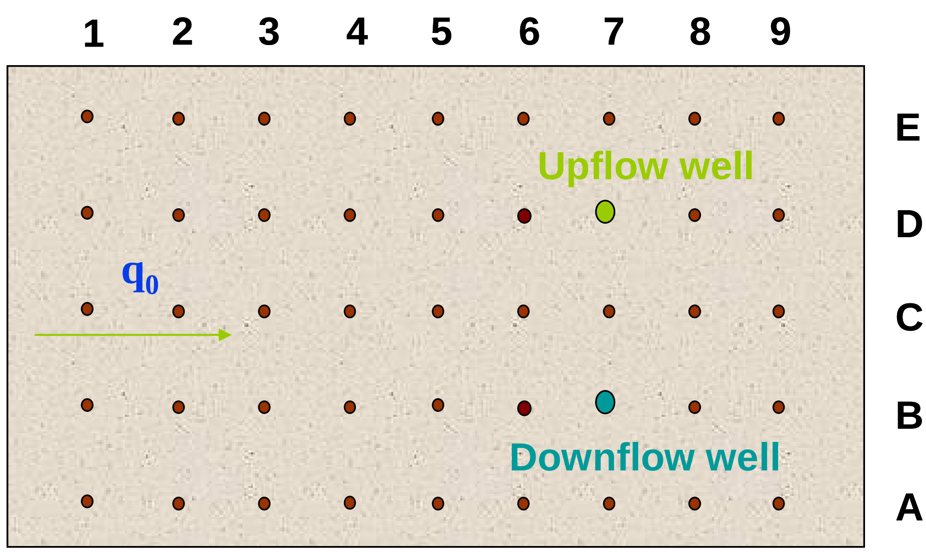

Figure 7. Plan view showing two TRWs

3.3.2 Multi-dipole technique experiments

After measuring water levels to establish the regional hydraulic gradient (which was determined to be 0.001319 for an aquifer flow of 2.8 m^3d^{-1}) the TRW pumps were operated. Steady-state drawdown at the downflow well and mounding at the upflow well was measured. Each well has 3 measurement points, the "top" (2.6 m from the bottom of the artificial aquifer), the upper screen (1.65 m above the bottom of the aquifer), and the lower screen (0.85 m above the bottom of the aquifer).

3.3.3 Tracer test technique experiments

TRWs were installed in the artificial aquifer at locations 7B and 7D (the upflow TRW at 7D, the downflow at 7B). The injection screens (the upper screen of the upflow well and the lower screen of the downflow well) and the extraction screens (the lower screen of the upflow well and the upper screen of the downflow well) were constructed using 2.5 *cm* diameter PVC. The injection/extraction screens are 22.5 *cm* long, each consisting of two 7.5 *cm* long PVC slotted sections separated by a 7.5 *cm* long PVC blank. The injection and extraction screens in each well are separated by 1.28 *m*, with the upper and lower end of each screen isolated using inflatable rubber packers. Two pumps were used (one for each TRW) to extract water from the extraction screen and inject water into the injection screen at a specified flow rate.

After measuring the water levels at two piezometers, upgradient and downgradient, to calculate the hydraulic gradient (determined to be 0.00148 at an aquifer flow rate of 2.94 m^3d^{-1}), the TRW pumps were turned on. The downflow and upflow wells were operated at 2.56 m^3d^{-1} and 2.32 m^3d^{-1}, respectively. Bromide tracer was injected into the injection screen of the upflow well and nitrate injected into the injection screen of the downflow well. Injection of bromide and nitrate tracers was continued for 240 and 336 hours, respectively, until steady-state concentrations were reached at the two extraction screens. Concentrations of bromide, chloride, and nitrate were measured over time at all four TRW screens, for application of the tracer test technique.

The concentration of chloride at all screens averaged 10.48 mg/L.

3.4 IPT method experiment

For measuring mass flux using the IPT method, three experiments were implemented under different conditions. Experiment 1 was repeated in order to obtain a preliminary estimate of method precision. Table 1 shows the pumping and observation wells used in the three IPT experiments run in the artificial aquifer.

In IPT experiments 1 and 3, a single pumping well was used while in experiment 2 three pumping wells were used. In experiments 1 and 2 the pumping and/or observation wells were aligned perpendicular to the groundwater regional flow direction while in experiment 3 the observation wells were at an angle to the regional flow direction.

Table 1. Pumping and observation wells for IPT Experiments

Experiment		Pumping well	Observation well
1		3C	7B, 7C, 7D
2		2B, 2C, 2D	8B, 8C, 8D
3	0°	4D	7D
	26.6°		6C
	63.4°		5B

To obtain plots of Δh as a function of $\sum_{i=1}^{N} Q_i \ln \frac{d_{obs[i]}{}^2}{d_{w[i]}{}^2}$ all three IPT experiments were conducted with four pumping rates. To apply Equation (15) it is necessary that the plot of Δh as a function of $\sum_{i=1}^{N} Q_i \ln \frac{d_{obs[i]}{}^2}{d_{w[i]}{}^2}$ cross the x-axis (Δh = 0), so it is desirable

that pumping rates be chosen so that two of the pumping rates lead to positive values of Δh and two lead to negative values. Prior to running the tests, back-of-the-envelope calculations were accomplished to estimate the appropriate four pumping rates.

In each experiment, the pumps were started at the lowest pumping rate and kept running until steady-state water levels were reached. In this study, it was estimated that 18 hrs of constant pumping was adequate to achieve steady-state conditions in the artificial aquifer. After running the pumping well for 18 hours, the hydraulic head at the pumping well was observed for at least 1 hour, and if the water level remained constant, equilibrium conditions were assumed. After measuring the water levels of the pumping and observation wells at the lowest pumping rate, the rate was increased. This was repeated until all four pumping rates were run for each experiment.

IV. Results and Discussion

4.1 Introduction

In section 4.2, the data and flux measurement results obtained from TRW method experiments are presented and analyzed. In section 4.3, the data and results form IPT method experiments are presented and analyzed.

4.2 TRW method

4.2.1 Multi-dipole method

During the evaluation, the mounding (positive) and drawdown (negative) at the 3 measurement points for each TRW was measured (see Table 2).

Table 2. Drawdown (negative) and mounding (positive) at the TRWs for application of the multi-dipole approach

Downflow well (7B)				Upflow well (7D)			
Pumping rate (m^3/day)	Upper screen (mm)	Lower screen (mm)	Top (mm)	Pumping rate (m^3/day)	Upper Screen (mm)	Lower Screen (mm)	Top (mm)
1.47	-11.0	20	-3.6	1.49	23.5	-8.0	1.6
2.77	-14.5	28	-6.0	2.85	38.5	-17.0	3.0
4.35	-35.0	69	-8.8	4.22	58.5	-30.5	6.2
5.85	-67.0	93	-12.0	5.71	79.5	-46.0	9.6
7.11	-90.0	129	-14.4	7.19	98	-60.0	14.2

* Water flow rate through the aquifer: 2.8 m^3/day

As described earlier, a genetic algorithm (Carroll, 1996) was used to obtain the best fit value of hydraulic conductivity that maximized the objective function in Equation (4) for all five pumping rates.

Table 3. Hydraulic conductivities and mass fluxes measured using the multi-dipole approach

Pumping rate (L/min)		Hydraulic conductivity (m/d)			Mass fluxes (g/m^2*d)		
		Anisotropic ($k_r \neq k_z$)		Isotropic ($k_r = k_z$)	Measured		Actual
Downflow	Upflow	k_r	k_z	k	Anisotropic (using k_r)	Isotropic	
1.47	1.49	8.15	0.15	5.16	0.11	0.07	2.41
2.77	2.85	10.26	0.31	4.93	0.14	0.07	
4.35	4.22	4.63	0.07	3.81	0.06	0.05	
5.85	5.71	5.53	0.11	3.40	0.08	0.05	
7.11	7.19	4.52	0.05	3.30	0.06	0.05	
Using all data		4.56	0.05	4.68	0.06	0.06	

* k_r : Horizontal hydraulic conductivity, k_z : Vertical hydraulic conductivity

Table 3 shows the best fit values of hydraulic conductivity, chloride mass flux measured, and actual mass flux. The actual chloride mass flux of 2.41 g m^{-2}d^{-1} was determined by multiplying the chloride concentration of 10.48 g/m^3 by the flow through the aquifer (2.8 m^{3}d^{-1}) and dividing by the cross-sectional area of the aquifer (12.2 m^2). As shown in Table 2, the measured mass fluxes are one to two orders of magnitude less than the actual flux. It appears that the multi-dipole technique is insufficiently accurate

to be used to measure flux. Kim (2005) speculated that the inaccuracy was due to the sensitivity of method results to relatively small head measurements, and that increasing the TRW pumping rates would improve measurements. Results from this study, however, indicate that increased TRW pumping rates do not improve results, and method inaccuracies are due to some other problem.

4.2.2 Tracer test technique

TRWs were installed in the artificial aquifer at locations 7B and 7D (the upflow TRW at 7D, the downflow at 7B). The injection screens (the upper screen of the upflow well and the lower screen of the downflow well) and the extraction screens (the lower screen of the upflow well and the upper screen of the downflow well) were constructed using 2.5 *cm* diameter PVC. The injection/extraction screens are 22.5 *cm* long, each consisting of two 7.5 *cm* long PVC slotted sections separated by a 7.5 *cm* long PVC blank. The injection and extraction screens in each well are separated by 1.28 *m*, with the upper and lower end of each screen isolated using inflatable rubber packers. Two pumps were used (one for each TRW) to extract water from the extraction screen and inject water into the injection screen at a specified flow rate.

After measuring the water levels at two piezometers, upgradient and downgradient, to calculate the hydraulic gradient (determined to be 0.00148 at an aquifer flow rate of 2.94 m^3d^{-1}), the TRW pumps were turned on. The downflow and upflow wells were operated at 2.56 m^3d^{-1} and 2.32 m^3d^{-1}, respectively. Bromide tracer was injected into the injection screen of the upflow well and nitrate injected into the injection screen of the downflow well. Injection of bromide and nitrate tracers was continued for

240 and 336 hours, respectively, until steady-state concentrations were reached at the two extraction screens. Concentrations of bromide, chloride, and nitrate were measured over time at all four TRW screens, for application of the tracer test technique.

The concentration of chloride at all screens averaged 10.48 mg/L. Figures 8 and 9 show the concentration of bromide and nitrate, respectively, over time at the four TRW well screens.

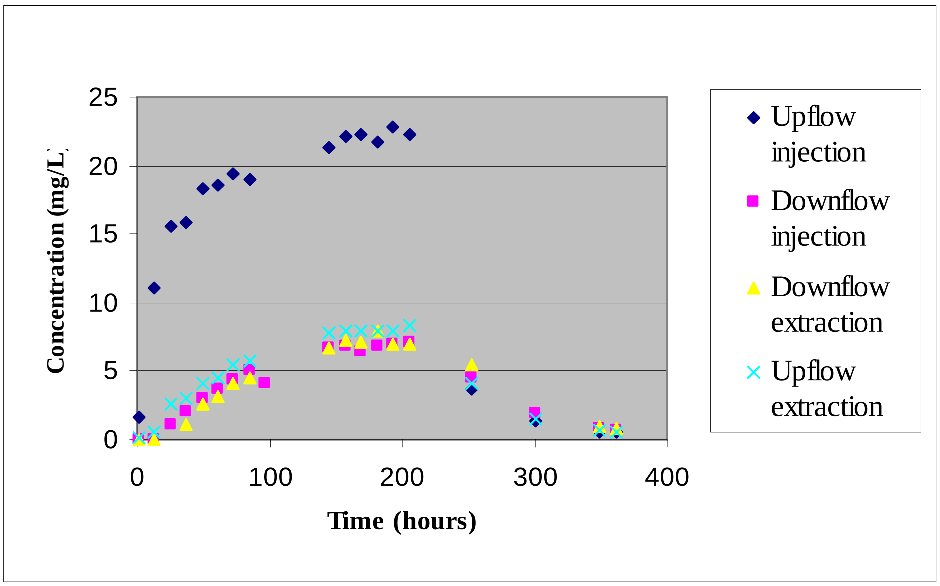

Figure 8. Bromide concentration over time at TRW screens

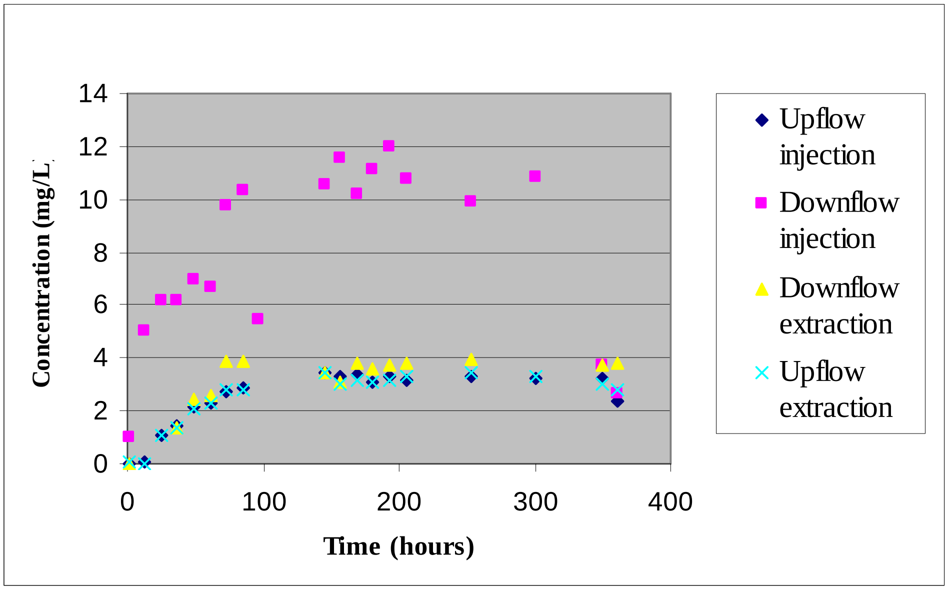

Figure 9. . Nitrate concentration over time at TRWs

Note that to apply equation (5) the steady-state tracer concentrations at the well screens are needed. As shown in Figure (8), the bromide concentration has reached steady-state at about 145 hours. Bromide steady-state concentration is obtained by averaging the measured concentrations from 145 to 205 hours. As shown in Figure (9), the nitrate concentration also has reached steady-state at about 145 hours. Nitrate steady-state concentration is obtained by averaging the measured concentrations from 145 to 301 hours. Table 4 lists the steady-state concentrations of tracers at the TRW's four screens. Kim (2005) used four different methods to estimate steady-state concentrations over different time ranges and found that the results were not sensitive to the estimation method.

Table 4. Steady–state tracer concentrations at TRW screens (g/m3)

Tracer	Upflow		Downflow	
	injection	extraction	injection	extraction
Bromide	22.10	7.94	7.00 (6.84)	7.00 (7.17)
Nitrate	3.26 (3.28)	3.26 (3.24)	10.87	3.63

* Note that according to the tracer test technique theory, bromide concentrations in the extraction and injection screens of the downflow well and the nitrate concentrations in the extraction and injection screens of the upflow well should be the same. Average values are used in this study. Numbers in parentheses indicate measured concentrations before averaging.

Perhaps the main disadvantage of the tracer test technique is the cost of tracers and their analysis. Kim (2005) proposed a cost-saving method based upon using a single tracer. If one assumes symmetry between the flow fields induced by each of the TRWs, it is possible to extrapolate the results of a test using a single tracer in order to apply the tracer test technique. If we assume symmetry, looking at Figure 4, we see I_{13} is equal to I_{42} and I_{12} is equal to I_{43}. Thus, the four unknowns in equation (5) are reduced to two unknowns, and it is only necessary to measure the steady-state concentrations of a single tracer at the four well screens to solve the two equations with two unknowns. Note that to apply this technique, it's also necessary to assume both TRWs are pumping at the same rate.

Table 5 shows the hydraulic conductivities and mass fluxes calculated using the tracer test technique. Values of hydraulic conductivity assuming anisotropy and isotropy were obtained by using a genetic algorithm (Carroll, 1996) to obtain the best fit

value of hydraulic conductivity that maximized the objective function in Equation (6). In the top row of Table 5 (the two-tracer method), results are presented based on the steady-state concentrations of both the bromide and nitrate tracers at the four well screens. The next four rows present results for the one tracer method described in the paragraph above. The actual chloride mass flux of 2.53 g $m^{-2}d^{-1}$ was determined by multiplying the chloride concentration of 10.48 g/m^3 by the flow through the aquifer (2.94 m^3d^{-1}) and dividing by the cross-sectional area of the aquifer (12.2 m^2).

Table 5. Hydraulic conductivities and mass flux calculated using the tracer test technique

<table>
<tr><th rowspan="4">Method</th><th rowspan="4">Tracer</th><th rowspan="4">Pumping rate (m^3/day)</th><th colspan="3" rowspan="2">Hydraulic conductivity(m/d)</th><th colspan="3">Mass flux (g/m^2*d)</th></tr>
<tr><th colspan="2">Measured</th><th rowspan="3">Actual</th></tr>
<tr><th colspan="2">Anisotropic ($k_r \neq k_z$)</th><th>Isotropic ($k_r = k_z$)</th><th rowspan="2">Anisotropic (using k_r)</th><th rowspan="2">Isotropic</th></tr>
<tr><th>k_r</th><th>k_z</th><th>k</th></tr>
<tr><td>Two tracers</td><td>Br-Nitrate</td><td>Upflow: 2.59 Downflow: 2.32</td><td>98.3</td><td>49.7</td><td>183.5</td><td>1.52</td><td>2.85</td><td rowspan="5">2.53</td></tr>
<tr><td rowspan="4">One tracer</td><td>Br</td><td>2.46</td><td>114</td><td>65.0</td><td>183.2</td><td>1.77</td><td>2.84</td></tr>
<tr><td>Nitrate</td><td>2.46</td><td>100</td><td>51.0</td><td>198.3</td><td>1.56</td><td>3.08</td></tr>
<tr><td>Br</td><td>2.59</td><td>97.7</td><td>50.9</td><td>188.1</td><td>1.51</td><td>2.92</td></tr>
<tr><td>Nitrate</td><td>2.32</td><td>98.2</td><td>50.8</td><td>187.1</td><td>1.52</td><td>2.90</td></tr>
</table>

For the two-tracer test assuming isotropy, the measurement overestimates the actual flux by only 13 %. For the one tracer test assuming isotropy, the measured mass fluxes are also close to the actual value, overestimating the actual value between 13% and 22%. It appears that at least in the relatively homogeneous conditions of the artificial aquifer, the assumption of symmetry is appropriate and results obtained from a single

tracer approximate the results obtained using two tracers. Assuming anisotropy, the mass flux measurements were lower than those assuming isotropy, underestimating the actual value between 30% and 40%. It appears that for the relatively homogeneous and isotropic artificial aquifer, the mass fluxes measured by the tracer test technique when assuming isotropy are better than those measured assuming anisotropic conditions. Similarly, Kim (2005) found that for the artificial aquifer, results obtained when assuming isotropy were significantly more accurate than were obtained assuming anisotropy.

4.3 IPT method

Table 6, 7 and 8 show the measurements of the hydraulic head at each pumping and observation well at all pumping rates for Experiments 1, 2, and 3 respectively. To apply the IPT method, the regional flow direction must be determined. The regional flow direction can be determined by head measurements with the pumps turned off. The coordinate system is set up with the pumping well at the origin. In the case of multiple pumping wells (Experiment 2), the center well is located at the origin and the other wells are aligned on the y-axis. The x-axis is defined as the line connecting the pumping well at the origin with an observation well. In the case of Experiments 1 and 2, the x-axis was the line connecting the pumping well at 3C with observation well 7C (experiment 1) or the line connecting the pumping well at 2C with the observation well at 8C (experiment 2). In both cases, the x-axis and regional groundwater flow direction coincided, so α in Equation (6) was set equal to 0.

Table 6. Measurements of hydraulic head for IPT experiment 1

Pumping rate (L/min)	Hydraulic head (mm)			
	Pumping well	Observation well		
	3C	7B	7C	7D
0	109.8	100	100	100
0.45	108.2	99.4	99.8	99.6
2.11	95.2	96.4	96.6	96.4
2.90	87.4	93.8	93.6	93.0
3.44	82.4	92	92.2	92.4

Table 7. Measurements of hydraulic head for IPT experiment 2

Pumping rate at each well (L/min)	Hydraulic head (mm)					
	Pumping well			Observation well		
	2B	2C	2D	8B	8C	8D
0	·	115	·	100	100	100
0.14	114.8	114.2	112.8	99.8	99.6	99.8
0.64	·	104.6	·	98.4	98.2	97.8
0.98	100.2	99.4	·	96.4	96.2	95.2
1.31	·	94.2	·	93.8	94.2	94.2

Table 8. Measurements of hydraulic head for IPT experiment 3

Pumping rate (L/min)	Hydraulic head (mm)			
	Pumping well	Observation well		
	4D	5B	6C	7D
0	105.0	102.6	100.0	98.0
2.0	91.6	96.2	94.8	93.0
2.5	87.4	94.6	93.0	91.4
3.0	84.0	93.0	91.4	90.2
4.18	74.8	87.6	87.4	86.8

For experiment 3, where the pumping well was at 4D, the value of α was 0.464 radians (26.6°), and 1.11 radians (63.4°) for the observation wells at 7D, 6C, and 5B, respectively. For the three experiments, the Δh vs $\sum_{i=1}^{N} Q_i \ln \frac{d_{obs[i]}^{2}}{d_{w[i]}^{2}}$ plots are shown in Figures from 10 to 14. Note that in accordance with the theory, the plots are relatively linear, with correlation coefficients close to 1.0. The fact that the study was done in a relatively homogeneous confined artificial aquifer undoubtedly contributed to the linearity of the results.

Using equation (15), the intercept of the x-axis in Figure 10 can be used to derive the Darcy velocity (q_0). Multiplying Darcy velocity by the concentration gives us an estimate of flux. Darcy velocities and flux measured by each experiment are shown in Table 9. The actual chloride mass flux was determined by multiplying the chloride concentration of 10.48 g/m^3 by the flow through the aquifer (3.75, 3.95, and 3.82 m^{3}d^{-1} for experiments 1, 2, and 3, respectively) and dividing by the cross-sectional area of the aquifer (12.2 m^2).

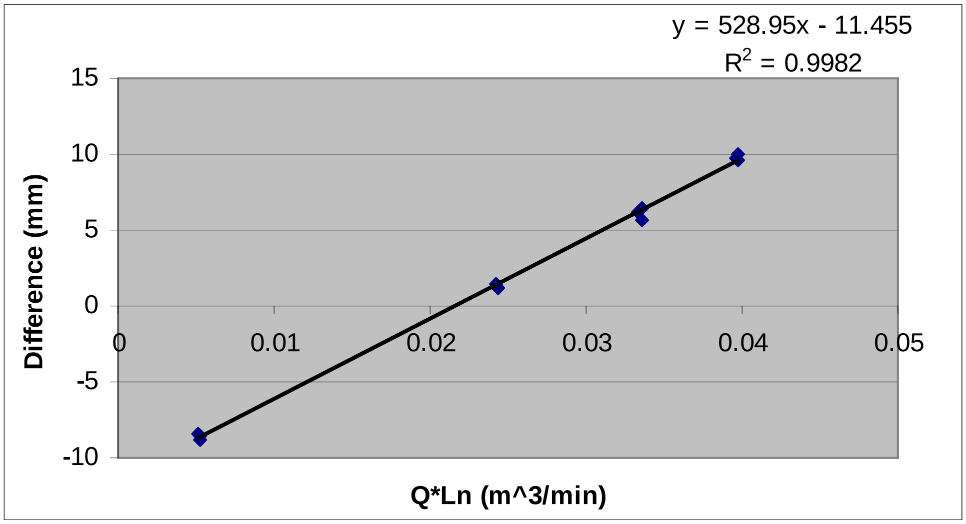

Figure 10. Plot to determine Darcy velocity for IPT Experiment 1

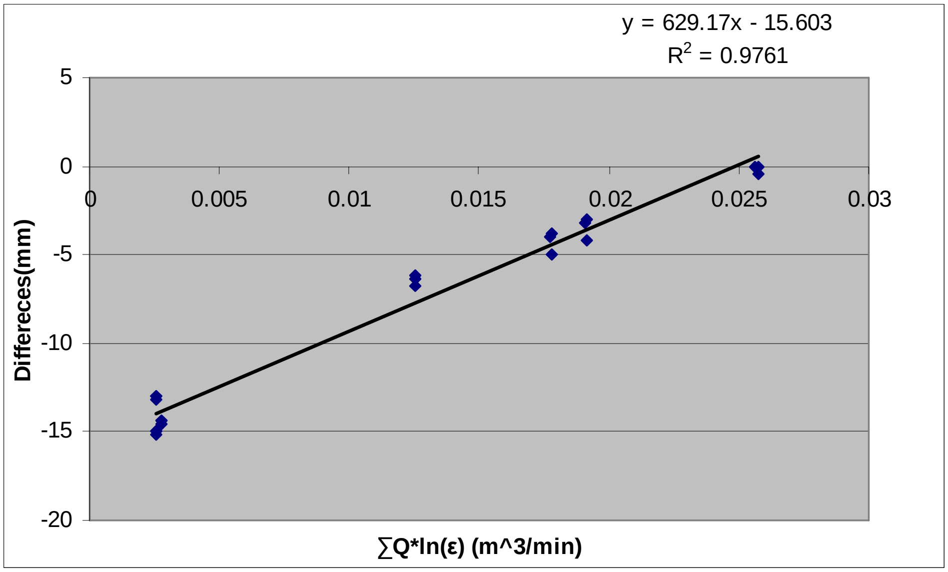

Figure 11. Plot to determine Darcy velocity for IPT Experiment 2

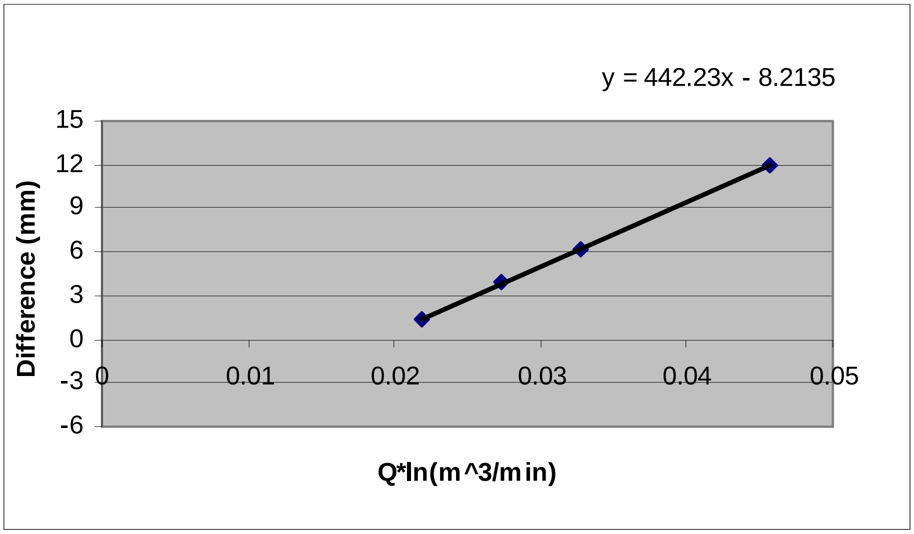

Figure 12. Plot to determine Darcy velocity for IPT Experiment 3 (α = 0°)

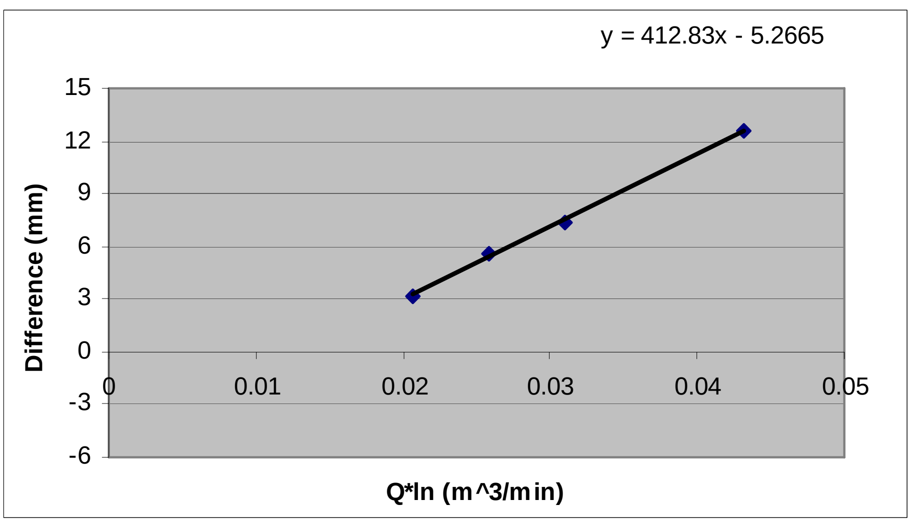

Figure 13. Plot to determine Darcy velocity for IPT Experiment 3 (α = 26.6°)

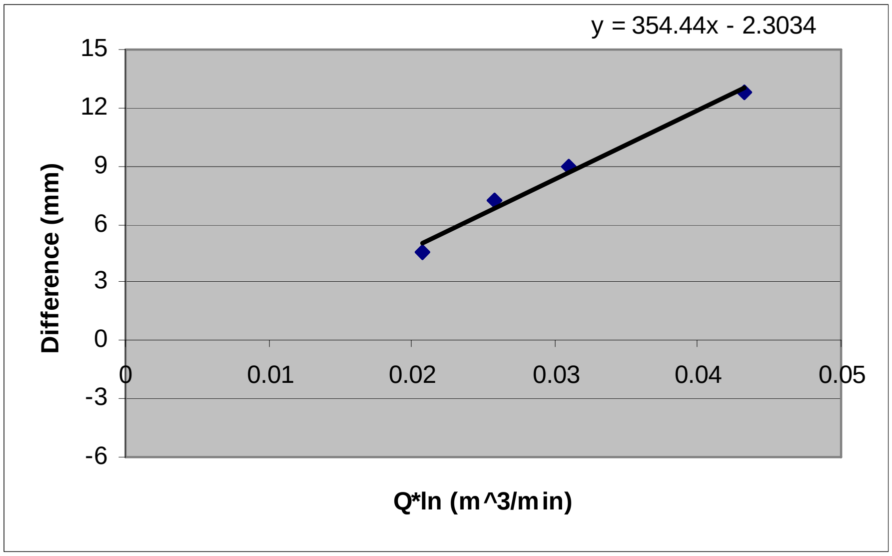

Figure 14. Plot to determine Darcy velocity for IPT Experiment 3 (α = 63.4°)

Table 9. Darcy velocity (q_0) and mass fluxes for IPT experiments

Experiment		∑Q*ln(ε) (Δh=0) (m^3/min)	q_0(m/day)	Mass flux (g/m^2*d)	
				Measured	Actual
1		0.022	0.24	2.51	4.64
2		0.025	0.18	1.91	4.89
3	0°	0.018	0.28	2.90	4.72
	26.6°	0.013	0.28	3.00	
	63.4°	0.006	0.29	3.00	

Note from Table 9 that the measured mass flux underestimates the actual flux by between 36% and 60%. This large an error is somewhat surprising, given the relative homogeneity of the artificial aquifer. We can consider several possible sources of error. There are a number of assumptions upon which the IPT method is based. The method

assumes the IPT is conducted in a confined aquifer, with infinite boundary conditions, uniform regional flow, and hydraulic heads are at steady state. Clearly, the artificial aquifer is not an infinite system. In order to account for the no-flow boundary established by the walls of the artificial aquifer, image wells can be used, as shown in Figure 15. Table 10 shows the measured fluxes when accounting for the no-flow boundaries. It appears that the measured flux is more accurate by between 7% and 19% when accounting for the boundaries.

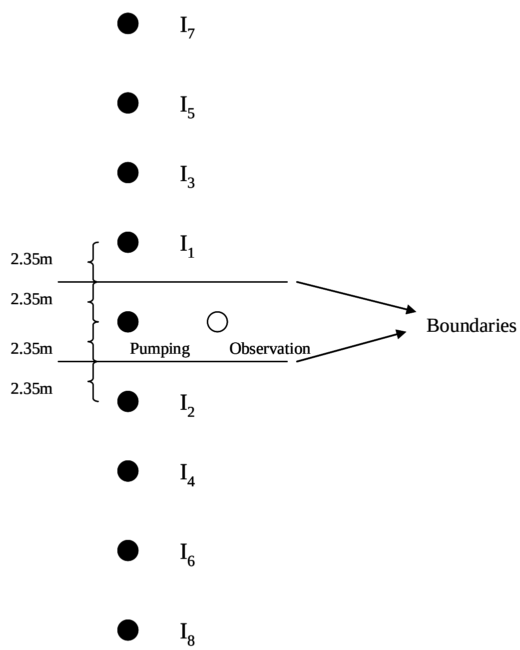

Figure 15. Image wells used to account for no-flow boundaries in IPT experiments

Table 10. Comparison between measured and actual mass fluxes for IPT experiments

Experiment		Mass flux (g/m^2*d)		
		Measured		Actual
		Without boundary effect	With boundary effect	
1		2.5	2.83	4.46
2		1.9	2.83	4.89
3	0°	2.9	3.2	4.72
	26.6°	2.9	3.2	
	63.4°	3.0	3.3	

Non-equilibrium conditions might also affect the accuracy of the IPT method. Unfortunately, the heads over time were not measured in this study. In order to check whether equilibrium was achieved, let us look at the measured drawdowns at the different pumping rates, and see if they are consistent with equilibrium conditions. At equilibrium, the difference in drawdown (Δs) between two wells at distances r_1 and r_2 from a well pumping at rate Q can be expressed by equation (16) (Domenico *et al.*, 1997).

$$s_1 - s_2 = \Delta s = \frac{2.3Q}{2\pi T}\log\frac{r_2}{r_1} \qquad (16)$$

From equation (16), we immediately see that

$$\frac{\Delta s_{Q_2}}{\Delta s_{Q_1}} = \frac{Q_2}{Q_1} \qquad (17)$$

where Δs_{Q_1} and Δs_{Q_2} are drawdowns at pumping rates Q_1 and Q_2, respectively. As

shown in equation (17), the ratio of Δs should be proportional to the ratio of pumping rates.

Table 11. Comparison of the ratio of pumping rates in IPT experiment 1 with the ratio of the difference in drawdown measured at pumping well 3C and observation well 7C

i	Q_i (L/min)	Δs_i (mm)	Ratio Q_i/Q_1	Ratio $\Delta s_i/\Delta s_1$
1	0.45	1.40	1.00	1.0
2	2.11	11.20	4.69	8.0
3	2.90	16.00	6.46	11.4
4	3.44	19.60	7.65	14.0

Table 11 compares the ratio of pumping rates in IPT experiment 1 with the ratio of the difference in drawdown measured at pumping well 3C and observation well 7C. From the table, we see that the ratios, which should be equal, differ by a factor of almost 2. Based on this, we suspect that we may not have achieved equilibrium.

Assuming the observation well had reached equilibrium at the lowest pumping rate of 0.45 L/min and that the pumping well had reached equilibrium at all pumping rates, but the head measurements at the observation wells at the higher pumping rates have not reached equilibrium, we can adjust the observation well heads according to the ratio of pumping rate Q. After adjusting the head measurements and recalculating, the measured mass flux for experiment 1 and 2 become 4.89 and 6.88 g $m^{-2}d^{-1}$, respectively, while the actual mass fluxes for the two experiments were 4.64 and 4.89 g $m^{-2}d^{-1}$, respectively (errors of 5% and 40%). Adjusting the observed heads in experiment 3 did

not affect the measured mass flux. Presumably, this is because the lowest pumping rate in experiment 3 was 2.0 L/min (as opposed to 0.45 L/min and 0.42 L/min for experiments 1 and 2, respectively), so the assumption that we are at equilibrium at the lowest pumping rate may be incorrect for experiment 3.

While the analysis above assumed that the artificial aquifer might not reach equilibrium after 18 hours pumping, a MODFLOW simulation showed this might not be a good assumption. In order to see how long the pumping well would have to be pumped to reach equilibrium, MODFLOW was run to simulate the conditions of experiment 1 with a pumping rate of 2.11 L/min. The simulation showed that equilibrium at the observation well was reached after 21 seconds and 1.08 minutes assuming realistic storativities of 2.7E-4 and 2.7E-3, respectively. It appears that 18 hours should be more than adequate to attain equilibrium.

In order to check the equilibrium condition, experiment 1 was repeated. Based on the data of head measurements over time at the pumping well, it appeared that the pumping well reached equilibrium after 500 min (8.3 hours) at all pumping rates (see Appendix A, Figure 1 - 4).

Another assumption that could affect the measurement is that the aquifer is confined. When the TRWs are pumped at high rates, dewatering could occur so that the water level might go below the confining layer of the artificial aquifer and unconfined conditions would result. If the aquifer is dewatered, this might also lead to violation of our assumption of equilibrium, as the time required for a confined aquifer to reach equilibrium at a given pumping rate is much greater than the time required for a confined aquifer. The possibility of dewatering was investigated during the second run of

experiment 1, but dewatering was not observed.

Another source of error is measurement error. It is difficult to measure the head accurately because the differences of head being measured at each pumping rate are just a few mm (see Figure 6, 7, and 8). For example, a measurement error of Δh of just 2 mm could change the measured flux by 5%.

Measurement error can be analyzed by comparing the two runs of experiment 1. Appendix A shows the results of the second run of experiment 1. The measured mass fluxes for experiment 1 were 2.51 and 3.10 g $m^{-2}d^{-1}$ for the first and second runs, respectively. Using these duplicate measurements, the 90% confidence interval for the true value can be estimated using equation (18) (McClave *et al.*, 2001)

$$\bar{x} \pm t_{\alpha/2}\left(\frac{s}{\sqrt{n}}\right) \tag{18}$$

Where

$\bar{x}$ = average of values

$t_{\alpha/2}$ = t statistic having (n-1) degrees of freedom

s = standard deviation

n = number of samples

As a result, the 90% confidence interval for experiment 1 is from 0.94 to 4.67 g $m^{-2}d^{-1}$. That is, we can say with 90% confidence that the true mass flux for experiment 1 falls in between 0.94 and 4.67 g $m^{-2}d^{-1}$. We see that the 90% confidence interval includes the actual value of 4.64 g $m^{-2}d^{-1}$.

V. Conclusions

5.1 Summary

In recent years, investigators have proposed contaminant mass flux as a critical measurement needed to support decision making at contaminated sites. Methods of measuring contaminant mass flux are being developed, and need to be validated. Two innovative approaches, the TRW and IPT methods, have been suggested to measure the mass flux. In this study, measurements from these two methods were compared with known fluxes in an artificial aquifer.

5.2 Conclusions

Results from using TRWs with the multi-dipole technique show that the measured mass fluxes were one or two orders of magnitude lower than the actual flux, and the technique appears to be not useable. Results of the tracer test technique show promise, with measurements within 15% of actual fluxes. Also encouraging was the fact that, at least in an artificial aquifer, the more inexpensive single tracer approach was approximately as accurate as the approach that used two tracers. The IPT method also shows promise. While measured fluxes underestimated the actual flux by at least 36%, it appears that errors may be reduced when one accounts for potential violations of method assumptions (infinite homogeneous confined aquifer, equilibrium conditions).

5.3 Recommendations

Based on the potential of the TRW method using the tracer technique, further

investigation is warranted. At Canterbury, New Zealand is a second facility that was constructed as a heterogeneous artificial aquifer. The TRW method can be validated in this second facility, to see how accurate it is under more realistic conditions of aquifer heterogeneity. In addition, replicate TRW experiments to allow for a more rigorous statistical analysis should be conducted.

Further investigation of the IPT method is needed in the homogeneous aquifer. Replicate experiments to allow for a more rigorous statistical analysis should be conducted, and the validity of method assumptions assessed. Follow-on studies should focus on developing procedures to help assure method assumptions are satisfied.

Appendix A. Results of IPT experiment 1 repeated

Table 1. Measurements of hydraulic head for IPT experiment 1 repeated

Pumping rate (L/min)	Hydraulic head (mm)	
	Pumping well	Observation well
	3C	7C
0	110.2	100.0
0.41	108.0	99.2
1.94	97.2	96.0
2.86	91.2	93.8
3.28	89.4	93

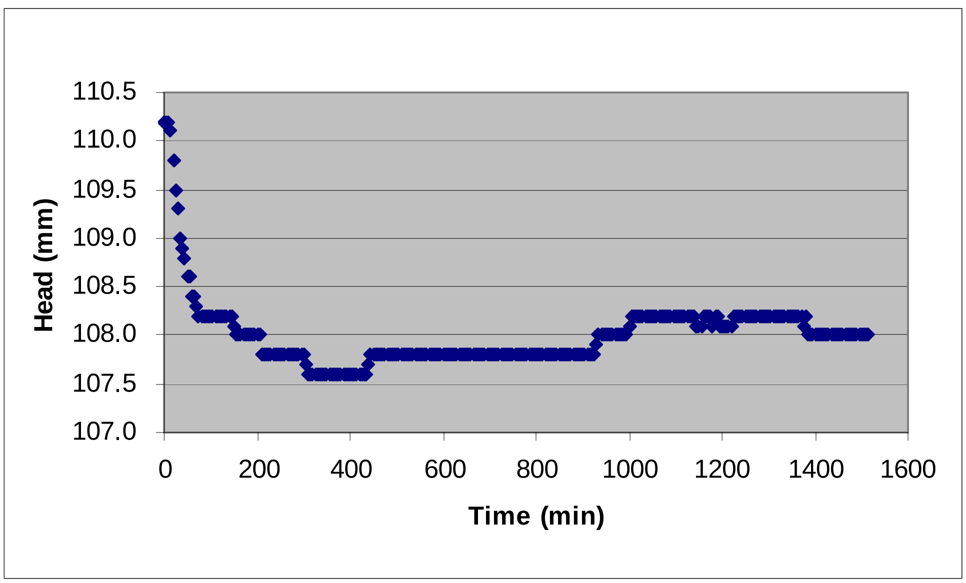

Figure 1. Measurements of hydraulic head over time at pumping rate 0.41 L/min

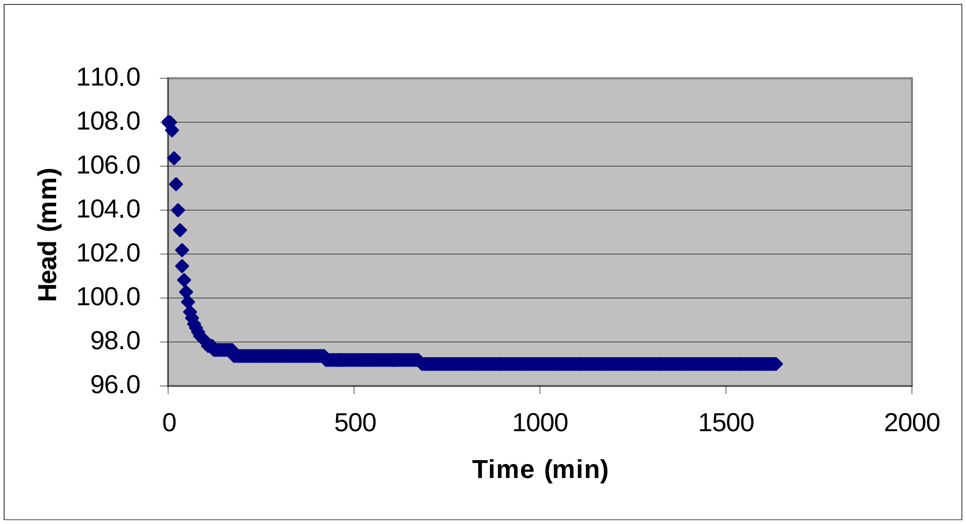

Figure 2. Measurements of hydraulic head over time at pumping rate 1.94 L/min

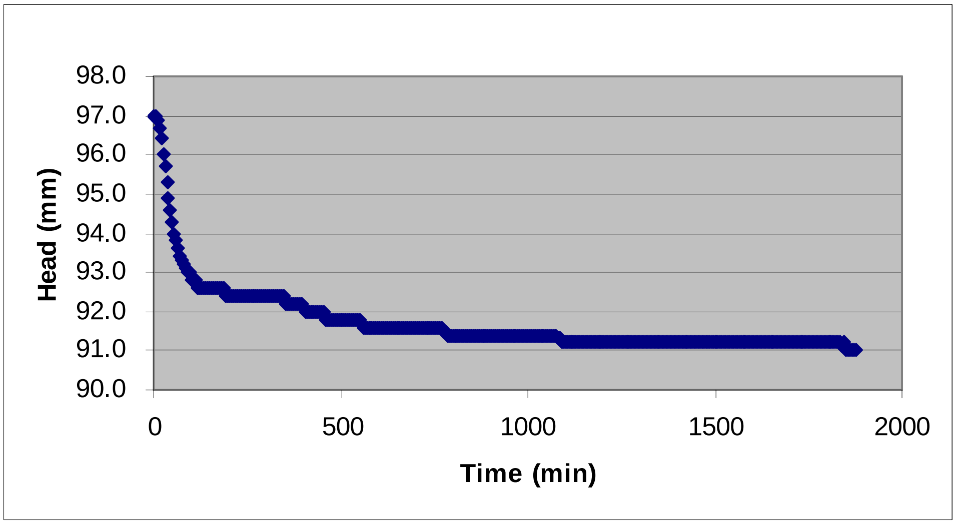

Figure 3. Measurements of hydraulic head over time at pumping rate 2.86 L/min

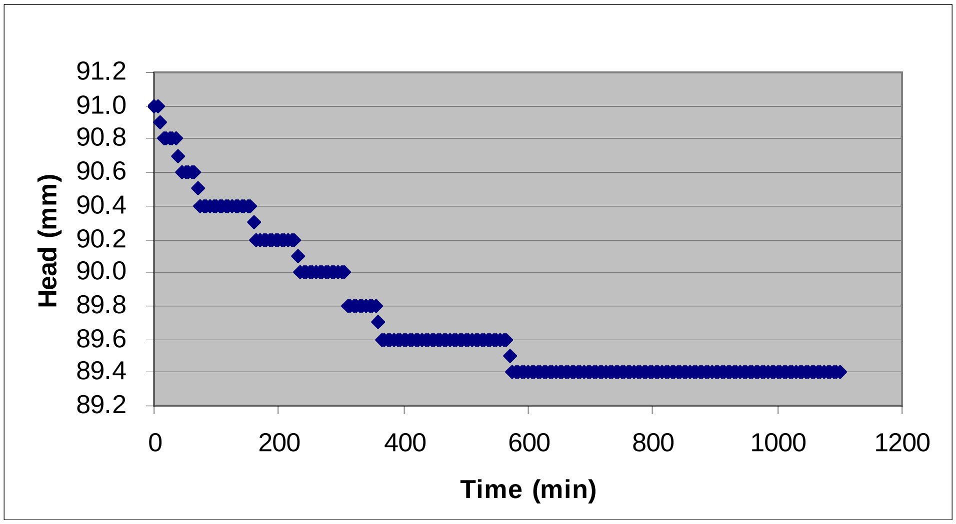

Figure 4. Measurements of hydraulic head over time at pumping rate 3.28 L/min

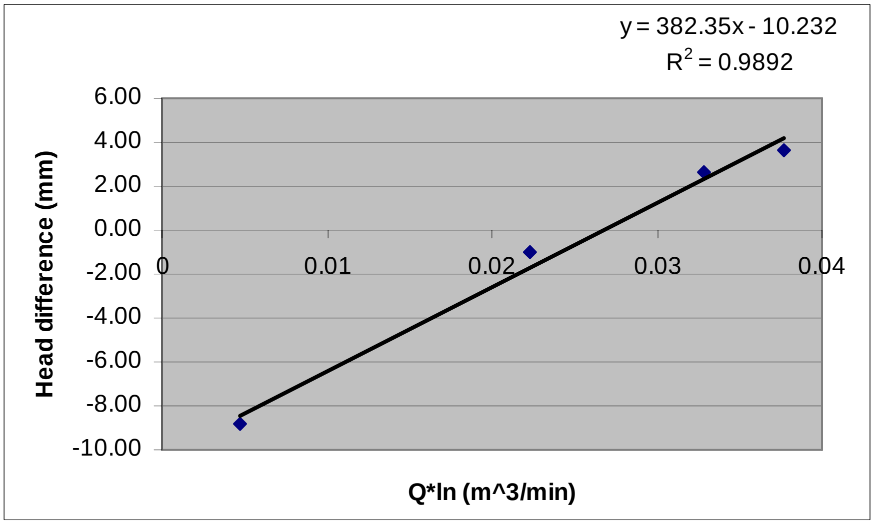

Figure 5. Plot to determine Darcy velocity for IPT Experiment 1 repeated

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Vita

Captain Hyouk Yoon graduated from Dae-shin High School in Dae-jeon, Republic of Korea in 1993. He entered Korea Military Academy (KMA) where he received the Bachelor of Science in Chemistry. Upon graduation, he received the commission of 2^{nd} Lieutenant, Army Infantry Officer.

He successfully performed various assignments in all around Korea for ten years. In August 2003, he entered the Graduate School of Engineering and Management, Air Force Institute of Technology. Upon graduation, he will be assigned to the Combined Forces Command (CFC) at Yong-san, Seoul.

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